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Sample dependence of the electron–electron scattering resistivity of copper whiskers

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Abstract. A calculation is presented of the size dependence of the normal electron–electron scattering and the Umklaap electron–electron scattering contributions to the electrical resistivity of copper whiskers. We find that the normal scattering term is larger than the Umklapp scattering term when the diameter of the copper whisker is smaller than about 10 μm. These results are in good agreement with recent measurements.

1. Introduction

In the past decade there has been considerable interest, both experimental and theoretical, in the electron–electron scattering contribution $\rho_{ee}(T)$ to the electrical resistivity of thin wires and films of non-transition metals (Caplin *et al* 1981, van der Maas 1983, Stesmans 1983, Yu *et al* 1984, 1989, De Gennaro and Rettori 1984, 1985, Kaveh and Wiser 1985, Thummes and Kotzler 1985, Zhao *et al* 1988, Gurzhi *et al* 1989a, b, Movshovitz and Wiser 1990a, b, 1991a, b, Qian *et al* 1991).

In the present study of $\rho_{ee}(T)$ for thin wires, we have calculated the diameter dependence of the contribution to $\rho_{ee}(T)$ due to normal electron–electron scattering (NEES) and that due to Umklapp electron–electron scattering (UEES). In order to compare our results with the recent measurements of $\rho_{ee}(T)$ for copper whiskers (Sprengel and Thummes 1990), we carried out the calculations using parameters appropriate to copper. We find, for example, that for pure copper wires thinner than about 10 μm, most of $\rho_{ee}(T)$ arises from NEES, whereas for thicker wires, UEES makes the major contribution.

The recent University of Hamburg data for $\rho_{expt}(T)$ for thin whiskers of copper (Sprengel and Thummes 1990), together with their previously reported data (Thummes and Kotzler 1985), constitute a set of measurements of $\rho_{expt}(T)$ at low temperatures for a series of copper whiskers whose diameters span a factor of three. Our calculated results are in good agreement with these data.

In section 2, the data are presented for the low-temperature resistivity of thin copper whiskers. The calculations of the diameter dependence of the NEES and UEES contributions to $\rho_{ee}(T)$ are described in section 3. In section 4, comparison is made between theory and experiment. The summary follows in section 5.

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2. The low-temperature resistivity data

For bulk samples of the non-transition metals at low temperatures (below about 1.5 K for copper), $\rho_{\rm expt}(T)$ varies quadratically with temperature,

$$\rho_{\rm expt}(T) = A_{\rm expt}T^2 \tag{1}$$

which is to be associated with electron–electron scattering. Therefore, the low-temperature data for $\rho_{\rm expt}(T)$ are summarized by stating the value for $A_{\rm expt}$.

Experimentally, it is found that the quadratic temperature dependence of $\rho_{\rm expt}(T)$ is retained for thin whiskers of copper (Thummes and Kotzler 1985, Sprengel and Thummes 1990). It should be noted that this result is not necessarily obvious. For potassium, for example, the temperature dependence of $\rho_{\rm expt}(T)$ for thin wires below 1 K is *not* quadratic (Yu *et al* 1984, Zhao *et al* 1988), even though bulk samples of potassium do satisfy equation (1).

For copper, the temperature dependence of $\rho_{\rm expt}(T)$ satisfies equation (1) below 1.5 K even for thin whiskers. However, the value of $A_{\rm expt}$ increases quite markedly as the diameter of the whisker decreases. The measured values of $A_{\rm expt}$ for whiskers of different diameters d are presented in table 1. All the measured whiskers originated from the same batch of pure copper, which was found experimentally (Thummes *et al* 1985) to correspond to a bulk mean free path for electron–impurity scattering of $\lambda_{\rm imp} = 240\ \mu$m. Since $\lambda_{\rm imp} \gg d$ for all the whiskers (see table 1), it follows that electron–surface scattering dominates the residual resistivity. As we shall see in the next section, this is the reason that the value of $A_{\rm expt}$ for the whiskers increases so sharply with decreasing thickness.

Table 1. Experimental $A_{\rm expt} = \rho_{\rm expt}(T)/T^2$ and the diameter d for each of the measured copper whiskers and for a bulk sample of copper. All whiskers had a bulk mean free path for electron–impurity scattering of $\lambda_{\rm imp} = 240\ \mu$m. The references for the data are denoted ST for Sprengel and Thummes (1990), TK for Thummes and Kotzler (1985) for the whiskers and SRS for Steenwyk *et al* (1981) for the bulk sample.

Diameter (μm)	$\lambda_{\rm imp}/d$	$A_{\rm expt}$ (fΩ cm K^{-2})	References
6.9	34.8	179	TK
10.3	23.3	130	ST
13.2	18.2	70	ST
19.1	12.6	51	ST
22	10.9	49	TK
$\sim$ 1500	$\ll 1$	27	SRS

3. Normal scattering and Umklapp scattering

3.1. Input data

The basic input data required for the calculation of the thickness dependence of the NEES and UEES contributions to $\rho_{\rm ee}(T)$ are the corresponding bulk electron mean free paths, denoted by $\lambda_{\rm NEES}(T)$ and $\lambda_{\rm UEES}(T)$, respectively. It is convenient to express these mean free paths in 'resistivity units' by defining temperature-independent quantities,

$$A^{\lambda}_{\rm NEES} = mv_{\rm F}/ne^2T^2\lambda_{\rm NEES}(T) \qquad A^{\lambda}_{\rm UEES} = mv_{\rm F}/ne^2T^2\lambda_{\rm UEES}(T)\,. \tag{2}$$

Since UEES is a resistive scattering mechanism, $A^{\lambda}_{\rm UEES}$ is equal to the coefficient of the T^2 term in equation (1) for a bulk sample of pure copper. This quantity has been measured (Steenwyk *et al* 1981) and found to be

$$A^{\lambda}_{\rm UEES} = A_{\rm expt} = 27\ {\rm f}\Omega\ {\rm cm\ K}^{-2}\,. \tag{3}$$

The situation for NEES is more complicated. For a bulk sample of a non-transition metal, the total electron momentum is conserved at each NEES collision, which therefore does not degrade the electric current. As a result, NEES does not contribute at all to $\rho_{\rm ee}(T)$ for a bulk sample, and one refers to NEES as a *non-resistive* scattering mechanism. Since one cannot determine $A^{\lambda}_{\rm NEES}$ by measuring $\rho_{\rm expt}(T)$, indirect methods must be used. Kaveh and Wiser (1981, 1983) have shown that the resistivity data for strained thick wires of copper can be used to deduce the following value:

$$A^{\lambda}_{\rm NEES} = 2A^{\lambda}_{\rm UEES} = 54\ {\rm f}\Omega\ {\rm cm\ K}^{-2}\,. \tag{4}$$

Finally, the value of $\rho_{\rm ee}(T)$ for a thin wire depends on the purity of the sample, specifically on the bulk mean free path for electron–impurity scattering $\lambda_{\rm imp}$. We used $\lambda_{\rm imp} = 240\ \mu$m, because this value characterizes the measured copper whiskers, and we wish to compare our results with these data.

3.2. Umklapp electron–electron scattering

Since UEES is a resistive electron scattering mechanism, the calculation of the UEES contribution to $\rho_{\rm ee}(T)$ for a thin wire, denoted $\rho_{\rm UEES}(T)$, can be carried out using the method of Sambles and co-workers (Sambles and Elsom 1980, Sambles *et al* 1982). The Sambles *et al* (1982) expression for ρ for a thin wire of diameter d is

$$\begin{aligned}\frac{\rho_\infty}{\rho} &= 1 - \frac{12\lambda_{\rm imp}}{\pi d}\int_0^{\pi/2} {\rm d}\theta \cos^2\theta\sin^2\theta \int_0^{\pi/2} {\rm d}\psi \sin\psi\ (1-p)(1-I)/(1-pI) \\ I &= I(\theta,\psi) = \exp(-d\sin\psi/\lambda_{\rm imp}\sin\theta)\end{aligned} \tag{5}$$

where ρ_∞ and $\lambda_{\rm imp}$ are respectively the bulk resistivity and the electron mean free path for electron–impurity scattering.

The specularity parameter p characterizes the smoothness of the wire surface. The values $p = 0$ and $p = 1$ indicate that electron–surface scattering is, respectively, completely diffuse and completely specular. However, Sambles *et al* have emphasized that it is important to take account of the fact that p is not a constant, but rather p depends on the angle at which the electron strikes the wire surface. Their recommended expression is that derived by Soffer (1967),

$$p = \exp[-(4\pi\alpha\cos\Theta)^2] = \exp[-(4\pi\alpha\sin\theta\sin\psi)^2] \tag{6}$$

where Θ is the angle between the electron trajectory and the normal to the wire surface. The surface-roughness parameter α is the ratio of the root-mean-square surface roughness to the electron deBroglie wavelength.

To obtain $\rho_{\rm UEES}(T)$ from equation (5), one must include UEES in the bulk electron mean free path λ, which now has two contributions:

$$1/\lambda = 1/\lambda_{\rm imp} + 1/\lambda_{\rm UEES}(T)\,. \tag{7}$$

Since $\lambda_{\rm UEES}(T) \gg \lambda_{\rm imp}$, we may expand equation (7) to obtain

$$\lambda \simeq \lambda_{\rm imp} - \lambda_{\rm imp}^2/\lambda_{\rm UEES}(T)\,. \tag{8}$$

One then replaces $\lambda_{\rm imp}$ by λ in the expression for ρ given in equation (5) and expands the integral to first order in $1/\lambda_{\rm UEES}(T)$, as indicated in equation (8). The first term in the expansion yields the Sambles *et al* result for ρ (which is of no interest here), whereas the second term yields $\rho_{\rm UEES}(T)$. A straightforward, although tedious, Taylor expansion of the integrand in equation (5) gives the desired result:

$$\begin{aligned}\frac{\rho_{\rm UEES}(T)}{\rho^{\infty}_{\rm UEES}(T)} &= \frac{2}{\Lambda} - \frac{12}{\pi\Lambda^2}\int_0^{\pi/2} {\rm d}\theta\sin\theta\cos^2\theta\int_0^{\pi/2}{\rm d}\psi\sin^2\psi\left[1-(1-p)^2I/(1-pI)^2\right]\\ \Lambda &= 1-(12\lambda_{\rm imp}/\pi d)\int_0^{\pi/2}{\rm d}\theta\cos^2\theta\sin^2\theta\int_0^{\pi/2}{\rm d}\psi\sin\psi(1-p)(1-I)I/(1-pI)\end{aligned} \tag{9}$$

where the experimental bulk value for $\rho^{\infty}_{\rm UEES}(T)/T^2$ for copper has been given in equation (3). In the limit of a very thick wire, $d \gg \lambda_{\rm imp}$, it is readily seen that $\Lambda \rightarrow 1$ and $I \rightarrow 0$, and then $\rho_{\rm UEES}(T) = \rho^{\infty}_{\rm UEES}(T)$, as required.

The single unknown parameter in the above expression for $\rho_{\rm UEES}(T)$ is the surface-roughness parameter α in equation (6). We found that the reasonable value $\alpha = 2.2$ gives the best overall agreement between the calculated values and the data for $\rho_{\rm expt}(T)$ for the copper whiskers.

The integrals in equation (9) have been evaluated numerically to yield $\rho_{\rm UEES}(T)$. In figure 1, we plot the calculated values of $A_{\rm UEES} = \rho_{\rm UEES}(T)/T^2$ as a function of the diameter of the copper wire. For $d > 50$ μm, $A_{\rm UEES}$ is seen to be nearly equal to the experimental bulk value given in equation (1). For thinner wires, however, $A_{\rm UEES}$ increases significantly, becoming twice the bulk value for a wire of diameter $d = 16$ μm.

3.3. Normal electron–electron scattering

The calculation of the NEES contribution to $\rho_{\rm ee}(T)$, denoted $\rho_{\rm NEES}(T)$, is much more difficult than the corresponding calculation of $\rho_{\rm UEES}(T)$, because NEES is a non-resistive scattering mechanism. The details of this calculation have recently been given, both for a thin film (Movshovitz and Wiser 1990b) and for a thin wire (Movshovitz and Wiser 1991b). Nevertheless, it is useful to review the basic physics. As already stated, a NEES collision is non-resistive and does not contribute to $\rho_{\rm ee}(T)$ for a bulk sample, but for a thin wire, NEES *does* contribute to $\rho_{\rm ee}(T)$ by altering the direction of the electron trajectory. This drives the electron toward the wire surface, where it suffers a resistive electron-surface collision, thus yielding a non-zero (positive) value for $\rho_{\rm NEES}(T)$.

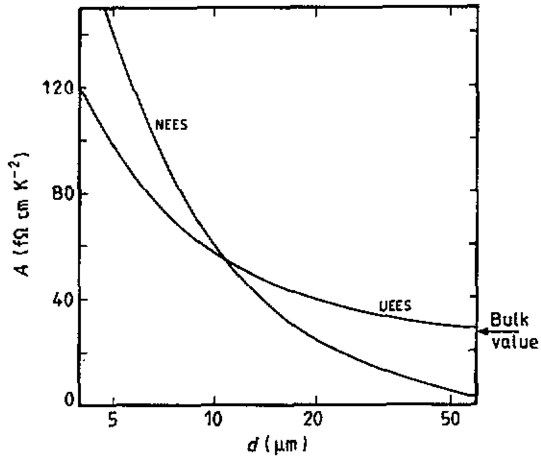

Figure 1. Thickness dependence of the NEES and the UEES contributions to the electrical resistivity of copper whiskers having a mean free path of $\lambda_{\rm imp} = 240$ μm for electron–impurity scattering. The measured bulk value of $A_{\rm expt} = 27$ fΩ cm K^{-2} is indicated by the arrow.

In principle, a NEES collision could also scatter an electron away from the wire surface, thereby increasing the mean free path between resistive electron–surface collisions and thus yielding a negative value for $\rho_{\rm NEES}(T)$. However, it is not hard to show (Movshovitz and Wiser 1991b) that for a pure thin wire, it is much more likely for NEES to scatter the electron *toward* the wire surface, implying a positive value for $\rho_{\rm NEES}(T)$. Moreover, the thinner the wire, the larger the magnitude of $\rho_{\rm NEES}(T)$.

The explicit expression for $A_{\rm NEES} = \rho_{\rm NEES}(T)/T^2$ has been found to be (Movshovitz and Wiser 1991b)

$$A_{\rm NEES} = \frac{A^{\lambda}_{\rm NEES}}{\Lambda^2}\left(\frac{8}{d^2}\int_0^{d/2} {\rm d}r\; rf^2(r) - \frac{12}{\pi}\int_0^{\pi/2} {\rm d}\theta\;\sin\theta\cos^2\theta\int_0^{\pi/2} {\rm d}\psi\sin^2\psi\frac{(1-p)I}{(1-pI)^2}\right) \tag{10}$$

where the quantities $A^{\lambda}_{\rm NEES}$ and Λ are given in equations (4) and (9) respectively, and the functions $p(\theta,\psi)$ and $I(\theta,\psi)$ in the integrand are given in equations (5) and (6) respectively. Finally, the function $f(r)$ is defined by

$$\begin{aligned} f(r) &= (3/\pi)\int_0^{\pi} {\rm d}\phi\int_0^{\pi/2} {\rm d}\theta\sin\theta\cos^2\theta\;(1-p){\rm e}^{-R/\lambda_{\rm imp}}/(1-p\,{\rm e}^{-S/\lambda_{\rm imp}}) \\ S &= S(r,\theta,\phi) = (d^2 - 4r^2\sin^2\phi)^{1/2}/\sin\theta \\ R &= R(r,\theta,\phi) = (r\cos\phi/\sin\theta) + S(r,\theta,\phi)/2\,. \end{aligned} \tag{11}$$

In the limit of a very thick wire, $d \gg \lambda_{\rm imp}$, it is readily seen that $f(r) \to 0$ and $I \to 0$, and then $A_{\rm NEES} = 0$, as required.

The integrals in equations (10) and (11) have been evaluated numerically to obtain $A_{\rm NEES}$. In figure 1, we plot the calculated values of $A_{\rm NEES}$ as a function of the diameter of the copper wire. For $d > 50$ μm, $A_{\rm NEES}$ is almost negligible, but for thinner wires,

A_{NEES} increases rapidly, being equal to the experimental bulk value at $d = 18\ \mu$m, and soon thereafter overtakes A_{UEES} at $d = 11\ \mu$m. For very thin wires, it is seen from figure 1 that A_{NEES} increases much more rapidly than does A_{UEES} with decreasing wire thickness.

4. Comparison between theory and experiment

The calculated value of $A_{\mathrm{ee}} = \rho_{\mathrm{ee}}(T)/T^2$ is the sum

$$A_{\mathrm{ee}} = A_{\mathrm{NEES}} + A_{\mathrm{UEES}} \tag{12}$$

where A_{NEES} and A_{UEES} are given in figure 1 for a copper whisker having $\lambda_{\mathrm{imp}} = 240\ \mu$m. In figure 2, we plot the calculted values of A_{ee} as a function of the diameter of the copper whisker. The symbols in figure 2 represent the measured values, with the circles and triangles referring to the data of Sprengel and Thummes (1990) and Thummes and Kotzler (1985) respectively. The overall agreement between theory and experiment is evident from the figure.

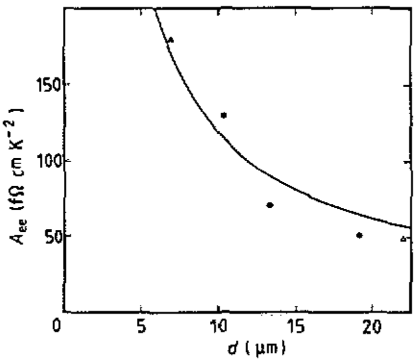

Figure 2. Thickness dependence of the coefficient A_{ee}. The symbols represent the data for A_{expt} for copper whiskers of differing diameters. The circles and triangles are the data of Sprengel and Thummes (1990) and Thummes and Kotzler (1985) respectively. The curve gives the calculated values of A_{ee}.

The $d = 22\ \mu$m whisker, indicated by the open triangle, was etched to roughen its surface. Therefore, for this whisker, a larger value of the surface-roughness parameter α is called for. However, using a larger value of α does not materially change the level of agreement between theory and experiment for this whisker. Even for $\alpha = \infty$, we obtain a calculated value of A_{ee} that is only 10% below the measured value, whereas for $\alpha = 2.2$, we find a value about 15% above the measured value, as shown in figure 2.

5. Summary

We have calculated the thickness dependence of both the normal electron–electron scattering and the Umklapp electron–electron scattering contributions to the electrical resistivity for thin wires of copper. Applying these results to the recent measurements of the low-temperature electrical resistivity of a series of copper whiskers leads to agreement between theory and experiment.

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